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(54) Title: HOT MELT ADHESIVE FOR ELASTIC BANDING AND METHOS FOR UTILIZING THE SAME

(57) Abstract

A purpose of the disclosed method is to impart gathers and elasticity to a relatively inelastic film, membrane, or web substrate, through elastic banding with a viscoelastic hot melt pressure-sensitive adhesive (PSA). Typically, the resulting elastic-banded substrate product (e.g. 40) will be cut into discrete units and formed into garments or body-encircling members such as disposable diapers. One step of the preferred method involves extruding a ribbon or band (13 or 113) comprising a viscoelastic hot melt PSA, which PSA has unusually high cohesion, stretchiness, and elasticity without excessive loss of adhesive bonding strength. (The viscoelastic behavior of the PSA is believed to be determined, at least in part, by the relative size of its crystalline domains and its rubbery domains and the glass transition temperatures and softening points of its components.) A second step of the preferred method involves bringing the band of hot melt (13 or 113) into adherent contact with a surface of a moving continuous substrate (22 or 32 or 132). A typical substrate would be the polyolefin film used in the manufacture of disposable diapers. The band can be bonded to the substrate through a pressure activation technique, wherein the band itself has the necessary inherent adhesive properties.

- 1 -

HOT MELT ADHESIVE FOR ELASTIC BANDING
AND METHOD FOR UTILIZING THE SAME

Technical Field

This invention relates to a method for imparting
5 elastic characteristics to materials which are
relatively inelastic through the use of a hot melt
pressure-sensitive adhesive. An aspect of this
invention relates to the formulation of a suitable
hot melt pressure-sensitive adhesive composition for
10 the aforementioned method. Still another aspect of
this invention relates to the manufacture of garments
or body-encircling members provided with an elastic
band. Additional aspects of the invention relate to
the elastic banding of such garments or body-encircling
15 members which are cut from a continuously moving
film, membrane, or web-like substrate and hot melt
pressure-sensitive compositions employed in the
elastic banding process which compositions can take
the form of extrudates, and elastic banding of discrete
20 articles wherein the hot melt pressure-sensitive
adhesive composition can be in the form of a tape.

Description of the Prior Art

In the garment industry, vulcanized rubber in
sheet or thread form is typically used for elastic
25 banding purposes. Traditionally, the vulcanized
rubber has been sewn, woven, or bonded to the garment
or discrete unit of material. Crosslinked synthetic
rubbers can be used in place of vulcanized natural
rubber for this purpose.

30   Sewing or weaving or similar attachment techniques
are not well suited to modern, high-production
processes and may even be inconvenient or cumbersome
to use in making homemade garments. A more efficient
technique for attaching an elastic band involves
35 the use of a heat-activated coating on the elastic
or a separate adhesive, which adhesive can be
coextensive in length with the elastic or merely
applied in spots. Taking his cue from this more

efficient approach, the patentee of U.S. Patent No.
4,081,301 (Buell), issued March 28, 1978 developed
a high-production process particularly well suited
to providing disposable diapers with elastic leg

5   bands. According to the Buell patent, glue applicators
can apply adhesive along the length of continuous
bands of elastic which are applied, in a stretched
condition, to the continuous web from which the
disposable diapers are made. The patent further

10   suggests that the elastic band or ribbon can be
coated with a heat-activated adhesive prior to contact
with the web. Still another suggestion relates to
the use of a heat-sealable elastic ribbon which can
be adhered to the web with the aid of a suitable

15   heating means.

Still greater efficiency (with a concurrent
simplification of the overall elastic banding
process) could theroetically be provided if the
adhesion of the elastic band to the substrate did not

20   require either a separate adhesive or a heating means.
Unfortunately, there are few guidelines in the
prior art for one who would attempt to formulate an
adhesive which could itself be an elastic band. It
is known that "pressure-sensitive" adhesives do not

25   require heat, solvents, moisture, or the like to
form a reasonably strong adhesive bond under normal
ambient conditions. It is also known that such
adhesives possess a degree of stretchiness, cohesion,
and elasticity as well as adhesion characteristics.

30   However, the four-fold balance of adhesion, cohesion,
stretchiness, and elasticity is a delicate one, and
any substantial increases in the last three of these
properties can result in unacceptable losses of
adhesion.

35   Since the invention of pressure-sensitive
adhesives, literally decades of r_search effort have

gone into the investigation of the aforementioned
four-fold balance and the development of tests for
reproducibly measuring the desired properties.   For
example, the adhesive bond strength of a pressure-
5   sensitive adhesive can be measured by 180° peel
resistance tests such as PSTC-1.   The adhesive tack
can be measured, for example, by probe tack tests
such as A.S.T.M. D2979.   Cohesion and stretchiness
of adhesives can be measured with modern tensile
10   testing equipment.

A particularly unusual problem may be
encountered when the adhesive is in a temperature
environment which is continuously above normal ambient
or room temperature.   It has been found that some
15   pressure-sensitive adhesives have their four-fold
balance significantly altered when the environment is
characterized by a modestly elevated temperature.
This finding is of great importance in the case
of garments or body-encircling members which are
20   continuously exposed to body temperature (e.g. 37° C.).

Most modern pressure-sensitive adhesives ("PSA's") are
applied to a substrate by one of three techniques:
coating from an organic-solvent based solution (e.g.
solvent casting), coating from a suspension or
25   dispersion such as an aqueous latex, and coating or
extruding of a hot melt pressure-sensitive composition.
The hot melt technique has a number of advantages;
for example, thicker layers of adhesive are readily
obtainable, solvent recovery is unnecessary, and
30   drying or "setting" time is minimal or nonexistent.
Research activity in the field of hot melt
pressure-sensitive adhesives has been very extensive,
and even a representative citation of references
drawn from this field would be difficult to provide.
35   The following selection of patents and literature is
believed to be reasonably representative.

U.S. Patent No.	Patentee	Issue Date
3,686,107	Russell	August 22, 1972
3,736,281	Russell	May 29, 1973
3,827,999	Crossland	August 6, 1974
5 3,935,338	Robertson et al	January 27, 1976
3,954,692	Downey	May 4, 1976
4,089,824	Bronstert et al	May 16, 1978

British Patent	Inventor	Publication Date
1,405,786	Crossland	September 10, 1975

10   Kirk-Othmer Encyclopedia of Chemical Technology,
2nd Edition, Volume 1, John Wiley & Sons, Inc., New
York, New York, 1963, pages 381-384.

"SOLPRENE® 418 in Pressure Sensitive Adhesives",
Bulletin 304 of Phillips Chemical Co., a division of
15 Phillips Petroleum Company.

Summary of the Invention

It has now been found that a greatly simplified
method for imparting elasticity to a relatively
inelastic substrate can be provided if one extrudes
20 a band comprising a suitable viscoelastic hot melt
pressure-sensitive adhesive, cools the thus-extruded
band to a temperature below its softening point
but above its glass transition temperature, and
brings the thus-cooled band into contact with the
25 substrate to form a pressure-sensitive adhesive (PSA)
bond, typically by means of pressure only, although
modest amounts of heat can be used also, if desired.
The band of hot melt can be pre-extruded and formed
into a convoluted roll of hot melt pressure-sensitive
30 adhesive tape. If the pre-extruded technique is used,
a cooling step is ordinarily unnecessary, since the
roll will ordinarily be stored and unreeled at
ordinary ambient temperatures. Extruding the band of
hot melt pressure-sensitive adhesive is by far the
35 .most practical approach when applying the elastic
band to a continuous non-elastomeric film, membrane,

or web substrate which is subsequently cut into
portions suitable for use as elastic-banded articles.
The convoluted tape approach is very practical when
a length of tape is to be applied to an individual
5   article.

In the context of this invention, a suitable
hot melt pressure-sensitive adhesive will have
viscoelastic behavior because of its glass transition
and/or softening point characteristics and
10 particularly because of a microstructure comprising
the combination of crystalline domains with
rubbery domains. The crystalline domains contribute
a pseudo-crosslinked character and greater elasticity
and cohesion. High elasticity and elastomeric
15 behavior are typically manifested by a storage
modulus (G') which is higher, and a loss tangent
(tan δ or G"/G') which is lower, than most conventional
PSA's. However, the storage modulus values cannot
be so high as to preclude viscoelastic behavior
20 in the temperature range of 25-50° C. A viscoelastic
solid, under stress, has some of the properties of
a highly viscous liquid (e.g. "creep" or "cold
flow") as well as some of the properties of an
elastomer. A PSA with suitable "creep" or "cold
25 flow" properties will have some tendency to flow in
the temperature range of 25-50° C., but this tendency
should be kept within limits, as manifested by a
limited range of loss modulus (G") values.

The following are considered to be illustrative
30 values for the G", G', and tan δ (tan $\delta = \frac{G"}{G'}$) of a

suitable PSA.

- 6 -

Property	*Values in 10^4 dynes/cm^2 at 25-50° C. and 0.01-0.25 Hz
Loss Modulus (G")	5 to 100*
Storage Modulus (G')	65 to 225*
Loss Tangent (tan $\delta = \frac{G"}{G'}$)	0.03 to 1.0 (no units)

5   Some frequency-dependence of these values can be
observed, but extreme temperature dependence is not
desirable. For example, a loss of more than
50×10^4 dynes/cm^2 in G' at 50° C., as compared to
25° C. indicates the likelihood of "heat set" or

10 inadequate elastomeric behavior at moderately elevated
temperatures. Permanent deformation due to elongation
should not exceed about 1.5 times the original length
of a sample of the PSA (i.e. a permanent increase in
length equal to 50% of the original length) throughout

15 the 25-50° C. range, using the dead load creep test
described subsequently.

Brief Description of the Drawing

Figure 1 is a schematic illustration of a typical
apparatus and typical method steps used in a preferred

20 embodiment of this invention.

Figure 2 is a similar schematic illustration of
another embodiment of this invention.

When used in this application, the following terms
have the indicated meanings.

25 "Pressure-sensitive adhesive" denotes those
adhesives which bond almost instantaneously when
contact pressure is applied to force the mating
surfaces together. Such adhesives have rather high
cohesive strength, such that, if the adhesive is

30 peeled away from a smooth surface to which it has
adhered, no apparent offsetting occurs and no
appreciable residue remains on the smooth surface.
True pressure-sensitive adhesives (PSA's) need not be

- 7 -

in a liquid or molten state in order to have adhesive
properties. Similarly, moisture, solvents, heat, or
the like are not needed to activate a PSA. Some
PSA's have aggressive tack or "quick stick" at room
5 temperature and tend to bond instantly upon contact.
The PSA's of this invention, on the other hand,
typically require light pressure to form a bond of
appreciable strength. In other words, the tack is
relatively non-aggressive and ordinarily would not
10 be sensed until some pressure were applied to the
surface of the PSA, causing it to "cold flow" in the
manner of most viscoelastic materials. Thus, a PSA
of this invention has a rheology which permits
sufficient flow under pressure to form a strong
15 adhesive bond while nevertheless maintaining a high
level of cohesion, stretchiness, and elasticity.

"Hot melt" refers to thermoplastic solids with
reasonably stable properties in the molten state,
which are easily melted at modestly elevated
20 temperatures (e.g. temperatures above 65° C.) and/or
easily extruded, and which can be melted and
resolidified a number of times without excessive
degradation of the thermoplastic properties. A
"hot melt pressure-sensitive adhesive" or "hot melt
25 PSA" refers to a hot melt adhesive having PSA
characteristics at temperatures below the softening
point and above the glass transition temperature of
the hot melt.

"Softening point" refers to a specific
30 temperature or range of temperatures which can be
determined by any of the standard softening point
tests such as the ring and ball ("R & B") test.
Accordingly, the term "softening point" includes
and subsumes "softening range".

35 "Elastomer" and "elastomeric" refer to a
material which, in the form of an unsupported film

- 8 -

or layer can be elongated to at least 100% of its
original length and which will return with force to
substantially its original length when permitted to
contract spontaneously. Thus, this invention
5 contemplates as "elastomers" those materials which
would be defined as "elastomeric" by the American
Society for Testing and Materials (A.S.T.M.).
"Non-elastomeric" materials are those which exhibit
some degree of significant deformation or "set"
10 when elongated 100% of their original length or
less; that is, such non-elastomeric materials typically
do not provide the elongation-resistant forces of
an elastomer.

"Essentially hydrocarbon resin" refers to a resin
15 in the molecular weight range of a few hundred up to
several thousand (e.g. 8,000) which is obtained or
synthesized from rather basic hydrocarbonaceous
materials such as petroleum, coal tar, turpentine,
olefins and other unsaturated simple hydrocarbons,
20 and the like. In the context of this invention, an
"essentially hydrocarbon resin" need not be a
hydrocarbon in the strictest sense of the term and
may contain oxygen, nitrogen, or sulfur, e.g. as
hetero-atoms or as atoms of functional groups.
25 Thus, an "essentially hydrocarbon resin" can be made
from a monomer such as coumarone (also known as
benzofuran). And, in industrial practice,
coumarone-indene resins are typically referred to
as "hydrocarbon resins".
30 The terms "loss tangent" (tan δ or G"/G'),
"storage modulus (G'), "loss compliance (J") and
"storage compliance" (J') are defined according to
established principles of dynamic mechanics. These
rheological quantities are measured on samples
35 approximately 2.5 mm in thickness placed between
25 cm parallel plate fixtures of a Rheometrics

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Mechanical Spectrometer (RMS). The sample was
allowed to equilibrate with the test temperature
(e.g. 25° C. or 50° C.). A minicomputer accurately
governs the application of a 5% peak-to-peak shear
5   strain to the sample. The frequency of the
application can be accurately controlled to a
fraction of a Hertz (Hz). The values of the complex
modulus (G*) and loss tangent are calculated by the
computer from geometry factors, peak-to-peak
10   amplitude of the torque signal, and phase lag of
the torque output wave. The definition of loss
tangent and the relationship between G*, G', and G"
provide two equations in two unknowns which can be
solved by the computer to provide G" and G', since
15   G* and loss tangent are both known values calculated
as described previously. The value for J' is given
by the reciprocal of G' divided by the expression
$1 + \tan^2 \delta$; the value of J" is given by the
reciprocal of G" divided by $1 + (\tan^2 \delta)^{-1}$. For
20   any of these values, the frequency in Hz (e.g.
0.25 Hz or 0.01 Hz) must be specified. Other
instruments for measuring these rheological properties
over a range of frequencies are known, e.g. the
"RHEOVIBRON".

25   The term "dead load deformation" or "dead load
creep" refers to a measurement of "cold flow" or
permanent deformation at one or more fixed test
temperatures, e.g. 23° C. or 25° C., 40 or 41° C., and
49 or 50° C. A sample of known length is suspended
30   vertically in a chamber maintained at the test
temperature and a weight (e.g. 1500 grams) is attached
to the lower (free) end of the sample. The
sample is cut to a size such that the force per unit
area is 1500 g/cm^2. After approximately 3 hours at
35   the test temperature, the sample is removed, the
weight is detached, and the sample is allowed to relax

- 10 -

under the influence of its own inherent elastomeric
forces. The length of the relaxed sample (L_2) is
compared to the original length (L_1) and the "dead
load creep" (permanent deformation) is determined
5   according to the formula (L_2-L_1)/L_1 x 100%.

Turning now to the Drawing, Figure 1 illustrates
the use of a pre-tensioned, cooled band of hot melt
PSA to bond two continuous substrates into an assembly
10 having gathers all along the bond line. A hot melt
reservoir 11 extrudes an elongated extrudate
(referred to herein as a "band") by forcing the hot
melt PSA material in reservoir 11 through an extrusion
die 12. The hot melt PSA band 13 comes into
15 contact with chill rolls 15 and 16 almost immediately
after the extrusion step, so that band 13 will be
cooled to a temperature below its softening point,
e.g. to a normal ambient temperature such as 20-25°
C.   (Typically, the hot melt PSA will be formulated
20 to have a glass transition temperature below normal
ambient temperatures.) Chill rolls 15 and 16, in
addition to cooling band 13, also advance it toward
tensioning rolls 17 and 18. Accordingly, the portions
23 and 33 of band 13 which are on either side of
25 tensioning rolls 17 and 18 will be under tension and
will be in an essentially elongated state.
Tensioning rolls 17 and 18 advance portion 33 of band
13 to nip rolls 27 and 28. Substrates 22 and 32 are
all the while being continuously unreeled from
30 storage rolls 21 and 31, so that the pretensioned,
cooled band of hot melt PSA 33 and substrates 22
and 32 all enter the nip provided by rolls 27 and 28
to be formed into the composite or assembled product
40 (i.e. the banded substrates). Although nip rolls
35 27 and 28 can be heated to a moderately elevated
temperature, in the preferred embodiment of this

- 11 -

invention, the modest pressure provided by the nip
rolls is all that is needed to adhesively bond
substrate 22 to substrate 32 with the hot-melt PSA
band 33. Since the nip rolls 27 and 28 provide only
5 line contact with the composite or assembly-comprising
substrate 22 and 32 and adhesive 33, it is difficult
to express the pressure applied by these rolls in
conventional terms such as Kg/m^2 or the like. Light
pressure on the order of tens or hundreds of grams
10 per square centimeter can be sufficient; however,
there is almost no upper limit on the pressure
applied by nip rolls 27 and 28 so long as the
rolls themselves or the substrates 22 and 32 are not
damaged. The hot melt PSA in band 13 can be formulated
15 to take into account the amount of pressure available
at nip rolls 27 and 28.

Since band 13 is a viscoelastic hot melt PSA
which meets the definition of an elastomer, it will
tend to contract spontaneously with force if the
20 tension applied to banded substrates 40 is less than
the tension on portion 33 of band 13. For example,
additional rolls or conveying devices (not shown)
can be used on banded substrates 40 merely to move
the composite product along toward a cutting station
25 and not exert any significant tension upon the banded
substrates. In such a situation, the portion 43
of band 13 on the exit side of nip rolls 27 and 28
will spontaneously cause the formation of gathers
41 all along the line of the adhesive bond between
30 band 43 and substrates 22 and 32.

The banded substrate product 40 can be cut into
individual elastic-banded articles such as disposable
diapers by techniques known in the art.

In the embodiment of the invention shown in
35 Figure 2, pre-tensioning of the hot melt PSA band 113
is not required. As in Figure 1, band 113 is extruded

from reservoir 111 through extrusion die 112. A
single substrate 132 is continuously unreeled from
storage roll 131. Substrate 132 is taken up by vacuum
chill roll 127 at a point which permits

5   pre-gathering of the substrate. Teeth-like projections
129 on the surface of vacuum chill roll 127 create
the flutes or gathers 141 in substrate 132. The two
chill rolls 127 and 128 serve to cool band 113 in
a manner analogous to the action of chill rolls 15

10 and 16 of Figure 1; in addition, these chill rolls
apply light pressure to the composite of the band
113 and the pre-gathered substrate 132, so that the
pressure-sensitive adhesive bond between band 113
and substrate 132 is formed without permanently

15 flattening out gathers 141. This result occurs
because, in the composite emerging from the exit side
of rolls 127 and 128, band 113 tends to be bonded
only to the peaks of gathers 141.

The foregoing methods are particularly well

20 suited to high-volume production techniques using
continuous substrates. For low-volume or batch
production, the elongation-resistant gathers can be
imparted to portions of the substrate by means of
a pressure-sensitive adhesive tape. In this technique,

25 the desired length of tape is simply unreeled from
a convoluted roll of hot melt pressure-sensitive
adhesive, which adhesive has been applied to a
flexible continuous backing. For good results in
this technique, the pressure-sensitive adhesive

30 tape should have an extruded layer at least 50
micrometers (μM) in thickness, more typically at
least 75 or 100 μM in thickness. If the flexible
continuous backing is relatively inelastic as compared
to the hot melt PSA layer of the tape, the backing

35 should have release characteristics, so that it can
be delaminated from the tape structure in the manner

of a transfer tape. The cohesive, self-supporting
PSA layer removed from the tape can be elongated and
applied to, for example, a garment in the stretched
condition. Gathers will be introduced into the garment
5 all along the lines of the adhesive bond after
pressure-sensitive bonding has been completed. A
modest amount of pressure can be applied by machine
or by hand to insure a strong adhesive bond. A
non-sticking coated roller or the like can be used
10 to apply the pressure, particularly superior non-stick
properties being obtained with fluoropolymer or,
less preferably, silicone coatings.

    Delamination of the tape is not necessary if the
backing of the pressure-sensitive adhesive tape is
15 itself highly elastomeric. Such a tape structure
can be provided, for example, by coextruding the
backing and the PSA through a single extrusion die.
The resulting stretchy tape can be rolled upon itself
or convoluted, particularly if the exposed side of
20 the elastomeric backing is treated so as to be
essentially non-sticky. Special sizings and the like
which prevent the formation of a PSA bond are known
in the art and can be employed for this purpose.

    Viscoelastic hot melt pressure-sensitive
25 adhesives (PSA's) of this invention preferably
comprise a rubbery block copolymer and at least
two different types of resins which associate with
different parts of the rubbery block copolymer molecule.
The resin which associates with the crystalline vinyl
30 arene end blocks of the block copolymer tends to
increase the size of these crystalline domains,
thereby, it is believed, decreasing the temperature
dependence of the elastic aspect of the viscoelastic
behavior of the PSA. (However, this invention is not
35 bound by any theory.) The adhesive character
of the PSA is believed to be

dependent in part upon the high-viscosity liquid
character of the PSA within the 25-50° C. temperature
range. A characteristic of a viscous liquid is
that it will yield to stress, and at least some strain
5   (in an oscillating stress situation) will be up to
90° out of phase with the stress. By contrast, in a
perfectly elastic solid the stress and strain would
always be in phase. The previously given broad
ranges of G", G', and loss tangent (and particularly
10   the preferred and optimum ranges given subsequently)
are believed to indicate a highly effective balance
of viscoelastic properties in the 25-50° C. range,
whereby at least some elasticity is provided (note
the loss tangent < 1 and the G' of 65×10^4
15   dynes/cm^2 or more), but in combination with some
ability to flow or "wet out" a substrate (note the
loss tangent > 0.03, the G' $< 200 \times 10^4$ dynes/cm^2,
and G" $< 100 \times 10^4$ dynes/cm^2), without resulting in
a viscosity so low as to permit excessive "creep"
20   or cold flow (note the G" $> 5 \times 10^4$).

        Although this invention is not bound by any
theory, it is believed that enlargement of
crystalline or vinyl arene domains will increase
the storage modulus (G') and decrease the dead load
25   creep, but some PSA behavior (e.g. peel strength)
may be lost. Conversely, enlargement of midblock
domains and/or increased tackifying of these domains
may increase PSA behavior but also increase dead
load creep and excessively decrease loss modulus
30   (G") and storage modulus (G') data. The ideal PSA
for this invention appears to provide a balance
of high-viscosity liquid and elastic solid behavior,
resulting in low dead load creep, good PSA
properties, and properly balanced rheological data.
35   The G', G", loss tangent, J' and J" data can be
considered to be "parameters" of the ultimate
properties of dead load creep and PSA characteristics.

A tackifying resin with aliphatic character and relatively minimal aromatic character can associate with the midblock of the block copolymer and, if properly selected, help provide this

5   viscoelastic balance in hot melt PSA behavior. Thus, a suitable viscoelastic hot melt PSA will typically comprise:

(1) a rubbery block copolymer which includes a rubbery midblock portion and which

10   is terminated with crystalline vinyl arene blocks,

(2) 20-150 parts by weight, per 100 parts by weight of said rubbery block copolymer, of a tackifying resin generally compatible with

15   and generally associated with the midblock portion of the block copolymer, and

(3) 10-150 parts by weight, per 100 parts by weight of the rubbery block copolymer, of an aromatic, essentially hydrocarbon resin

20   having a glass transition temperature and a softening point above those of the tackifying resin and the end blocks of the block copolymer, which aromatic resin is generally compatible with and generally associated with

25   the aforementioned end-blocks.

The hot melt pressure-sensitive adhesive will typically have a ball and ring softening point within the range of 65° to 240° C. It will exhibit elastomeric behavior above its glass transition temperature and

30   particularly for extended periods at body temperature (e.g. 37° C.).

In addition to the block copolymer and resins, the hot melt PSA can contain the usual antioxidants or stabilizers and essentially inert ingredients which

35   do not have a significant effect upon the properties of the combination of the rubbery block copolymer

and the resins. For example, minor amounts of fillers
and pigments can be included in the hot melt PSA,
tyipcally in amounts less than 5% by weight of the
total hot melt PSA composition. Substantially inert
5 extenders can also be included in the composition,
e.g. the typical hydrocarbon process oils. The amount
of process oil will typically also be kept below
5 weight-% of the composition, since large amounts
of oil can detract from the elastic recovery
10 characteristics of the PSA.

Typical antioxidants useful in PSA's of this
invention include the pentaerithritol phosphite
ester type (e.g. di[stearyl] pentaerithritol
diphosphite), the hindered phenol or polyphenol type,
15 and the like. Typical hindered phenol-type
antioxidants include those in which a phenolic
(i.e. hydroxyphenyl or hydroxybenzyl) group or
groups is or are substituted on a short hydrocarbon
chain, and the hydroxy group of the phenolic
20 substituent is hindered by nearby or adjacent alkyl
groups substituted on the phenol nucleus. Such
structures can be obtained, for example, by alkylating
or styrenating hydroxyphenyl compounds such as
phenols and cresols.

25   Typical pigments useful in formulating PSA's
of this invention include titanium dioxide,
typically having a particle size in the sub-micrometer
range, and similar finely divided materials.
Fillers may tend to be a bit coarser in particle
30 size, though still typically smaller than 40 µM
(minus 325 U.S. mesh), e.g. finely ground calcium
salts or silicates.

The following description of the preferred
ingredients of hot melt PSA's of this invention will
35 concentrate on the block copolymers and the resins.

A number of rubbery block copolymers can be
tackified to produce hot melt PSA compositions, as
can be seen from the disclosures in the aforementioned
U.S. Patent Nos. 3,686,107, 3,736,281, 3,827,999,

5   3,935,338, 3,954,692, and 4,089,824. Additional
disclosures of this type can be found in British
Patent 1,405,786 and trade literature of Phillips
Petroleum Company and Shell Chemical Company. The
block copolymers used in this invention are rubbery,

10   i.e. elastomeric. Though these copolymers are
thermoplastic in the sense that they can be melted,
formed, and resolidified several times with little
or no change in physical properties (assuming a
minimum of oxidative degradation), they exhibit some

15   of the characteristics of cross-linked or
vulcanized rubber. The apparent cross-linked
character is provided by the aforementioned
crystalline domains provided by vinyl arene terminal
blocks or end blocks. The block copolymers also

20   include a rubbery midblock portion which can be either
linear or branched. In typical examples of a branched
midblock, the midblock portion contains at least three
branches which can be radiating out from a central
hub or can be otherwise coupled together.

25   One way of synthesizing such rubbery block
copolymers is to begin with the polymerization of
the vinyl arene blocks which provide the end blocks.
Once the vinyl arene blocks have been formed, they can
be linked to elastomeric blocks, which elastomeric

30   blocks are typically obtained by polymerizing
unsaturated hydrocarbons, e.g. dienes such as
butadiene, isoprene, and dienes of higher hydrocarbons.
When an end block A is joined to an elastomer block B,
an A-B block copolymer unit is formed, which unit can

35   be coupled by various techniques or with various
coupling agents to provide a structure such as A-B-A,

which may in reality be two A-B blocks joined
together in a   tail/tail arrangement.
By a similar technique, a radial block copolymer
can be formed having the formula $(A-B)_n X$, wherein X
5   is the hub or central, polyfunctional coupling agent
and n is a number greater than 2. (If n were 2,
the polymer would be A-B-X-B-A, which is equivalent
to the A-B-A structure described previously and
is essentially linear.) Using the coupling agent
10   technique, the functionality of X determines the
number of A-B branches.

      Preferably, each block A has an average
molecular weight between 1,000 and 60,000, and each
block B has an average molecular weight between
15   5,000 and 450,000. The total molecular weight of
the block copolymer is preferably in excess of 100,000 or
200,000, e.g. 300,000. An extensive discussion of
rubbery radial block copolymers can be found in the
disclosure of the aforementioned U.S.Patent No.
20   4,089,824. As pointed out by the '824 patent,
the residual unsaturation in the midblock or
diene-containing portion of the block copolymer
molecule can be hydrogenated selectively so that the
content of olefinic double bonds in the radial
25   block copolymers can be reduced to a residual proportion
of less than 5% or even less than 2%. Such
hydrogenation tends to reduce sensitivity to
oxidative degradation and may have beneficial effects
upon elastomeric properties.

30   Preferred block copolymers used in this
invention have styrene end blocks and an isoprene
midblock portion. The isoprene typically comprises
the major amount of the repeating units in the
copolymer and can constitute, for example, 70% by
35   weight or more of the copolymer molecule. The
midblock, if branched, can have three or more branches,

and good results can be obtained with, for example,
four, five, or six branches. The midblock can be
hydrogenated, if desired.

5   Linear or A-B-A type block copolymers (including
A-B-A-B-A, etc.) are preferably selected on the
basis of end block content, large end blocks being
preferred. For S-I-S (styrene-isoprene-styrene)
block copolymers, a styrene content in excess of 14%
by weight is preferred, e.g. 15-30% by weight. A
10   commercially available example of such a linear
polymer is KRATON© 1111 rubber, an S-I-S polymer
which contains about 21.5% styrene units,
essentially the balance being isoprene units. Thus,
the optimum styrene content for linear S-I-S copolymers
15   appears to be greater than 20% by weight. As a
result of the higher styrene content, the polystyrene
end blocks have a relatively high molecular weight.
Typical properties of KRATON© 1111 are reported to
include a tensile strength of 2900 psi (2.0×10^6
20   Kg/m^2), a 300% modulus of 200 psi (1.4×10^5 Kg/m^2),
an elongation of 1200% at break, a 10% set at
break, and a Shore A hardness of 52; the Brookfield
viscosity of a toluene solution is 1300 centipoise
at room temperature, less than that of KRATON© 1107.

25   A variety of resins with tackifying properties
are compatible with polymerized diene polymer blocks,
including those diene blocks which have been
hydrogenated so as to become virtually identical,
chemically and physically, to polymerized mono-olefins
30   (e.g. polyethylene, polypropylene, polybutylene,
etc.). These "midblock" tackifying resins tend
to associate with the rubbery midblock of the linear
or radial block copolymer and thereby tend to extend
or build up as well as tackify these rubbery domains.
35   Both natural and synthetic "essentially hydrocarbon
resins" can be used as "midblock resins", provided

that these resins contain at least some aliphatic
character, which character can be provided by the
aliphatic portion of rosin acids, repeating isoprene
or other diene units (e.g. polymerized
5   1,3-pentadiene), polymerized cycloaliphatics, and the
like.

Although esters of polyhydric alcohols and rosin
acids will associate with a rubbery midblock, some
of these esters tend to detract from the elastic
10 recovery characteristics of the hot melt PSA and are
not preferred. It is generally more preferable to
use "essentially hydrocarbon resins", particularly
the so-called "terpene" resins, i.e. polymers with
repeating C_5H_8 or $C_{10}H_{16}$ units. These polymers can
15 be natural or synthetic and can be copolymers
(including terpolymers, etc.), since isoprene is an
olefin which can be copolymerized with other olefins.
Terpene-phenols have also been produced.

All terpene resins do not work with equal
20 effectiveness in this invention, and synthetic terpenes
having a softening point (ball and ring method) of
about 80 to about 115° C. are preferred, particularly
the commercially available resin known as "WINGTACK"
95. This commercially available terpene resin is
25 reported to be derived from a mixed olefin feedstock
as a by-product of isoprene or polyisoprene
production. According to U.S.Patent No. 3,935,338
and South African Patent No. 700,881, "WINGTACK" 95
(trademark of Goodyear Tire and Rubber Company) is
30 a thermoplastic tackifying resin essentially comprising
a copolymer of piperylene and 2-methyl-2-butene which
results from the cationic polymerization of 60%
piperylene, 10% isoprene, 5% cyclopentadiene, 15%
2-methyl-butene, and about 10% dimer. Other
35 tackifying resins of the same general type typically
comprise 20-80 weight-% of piperylene and 80-20
weight-% of 2-methyl-butene.

Good elastic memory or elastic recovery characteristics can be obtained with natural hydrocarbon resins such as "PICCOLYTE D-135" (trademark), a natural dipentene terpene resin.

5 However, this resin is not as effective as the "WINGTACK" 95 in providing good adhesive properties, e.g. good PSTC-1 peel.

The naturally occurring terpenes can be classified as monocyclic (dipentene), dicyclic (pinene), or

10 acyclic (micrene). A small amount of cyclic character is not detrimental in the context of this invention. A significant amount of aromatic character in the terpene resin is, however, ordinarily avoided, if such aromatic character is

15 sufficient to interfere with the midblock association properties of the resin.

As explained in British Patent No. 1,405,786, resins with aromatic character tend to associate with the vinyl arene end blocks. Such "end block"

20 resins include the coumarone-indenes, polystyrene, the polymethylstyrenes, the polyindenes, and other resins containing mono or polycyclic aromatic groups. Such resins are commercially available, e.g. as "PICCOTEX 75" (low molecular weight

25 alpha-methylstyrene-vinyl toluene synthetic copolymer), "PICCOTEX 100" (trademark for higher molecular weight version of "PICCOTEX 75"), "PICCOLASTIC D-150" (trademark for polystyrene resin), and the "CUMAR" resins (trademark for coumarone-indenes). It is

30 particularly desirable that the "end block" resin have a glass transition temperature and a softening point above those of the end block and of the "midblock" resin. For example, it would ordinarily not be desirable for the glass transition and for

35 significant heat softening to occur in the 80-110° C. range; hence, "end block" resins with somewhat

higher molecular weights and softening points above
115° C. are typically selected. From the standpoint
of strong elastic recovery (both initial and aged)
and good adhesive properties, the high softening

5 point coumarone-indene resins appear to be by far
the most effective. Such resins with softening
points within the range of 140-160° C. are commercially
available.

Considerable skill and knowledge already exist
10 in the PSA art with respect to determining
proportions of tackifiers and rubbery block copolymers.
However, it has been found that these proportions
cannot be selected with blind reliance upon prior
experience. Nor can one rely too heavily upon

15 PSTC-1 or probe tack values. Probe tack values and
initial (immediate) PSTC-1 values have been found
to be unreliable or unreproducible indicators of
performance in the context of this invention, whereas
PSTC-1 values taken 24 hours after the adhesive bond

20 has been formed are relatively reliable and
reproducible. "Dead load deformation" ("creep")
and the rheological properties (G', G", loss tangent,
J', and J") described previously have also been
found to be reliable, reproducible parameters of PSA

25 and elastomeric behavior.

Preferred PSTC-1 values determined 24 hours
after formation of the PSA bond at room temperature
on the standard steel plate using pressure from the
standard 2 Kg roller include values in excess of

30 1 pound per inch width (1.p.i.w.), i.e. more than 450
grams 180° peel force is required to delaminate a
tape/steel plate sample wherein the tape sample is
25.4 mm in width. PSTC-1 values in excess of
1500g/25.4 mm-sample or even 3000g/25.4mm-sample

35 can be obtained in practice. These values are
believed to indicate a reasonably permanent or
semi-permanent bond between a band of PSA and a

polymeric film substrate of the type used in disposable diapers.

Tensile strength values for the PSA at 20-25° C. can be determined as a measure of cohesive strength. Values in excess of 3×10^4 Kg/m^2 (e.g. above 33,000 Kg/m^2) can be obtained in practice. "Dead load deformation" values (1500 g/cm^2 for 3 hours at the test temperature) can be well below 100% and even below 50% ($\frac{L_2 - L_1}{L_1} \times 100\%$) throughout the range of 25° C. to 50° C. It is particularly desirable that this deformation remain roughly constant over this temperature range and preferably show a gain of less than 50% (e.g. less than about 35%) at 49° or 50° C., as compared to the room temperature value.

Preferred and optimum rheological parameters are set forth below.

TABLE OF PREFERRED AND OPTIMUM RHEOLOGICAL PROPERTIES
(All values in 10^4 dynes/cm^2 for G" and G' and in 10^{-8} cm^2/dyne for J' and J")

Property	PREFERRED (at 25-50° C.)		OPTIMUM (at 25-50° C.)	
	at 0.01 Hz	at 0.25 Hz	at 0.01 Hz	at 0.25 Hz
Loss Modulus (G")	5-25	10-100	8-16	15-35
Storage Modulus (G')	75-200	75-200	100-175	120-200
Loss Tangent (tan δ)	0.05-.3	0.08-1.0	0.07-0.10	0.01-0.25
Storage Compliance (J')	40-100	40-100	60-80	50-70
Loss Compliance (J")	2-20	5-40	5-8	5-15

The temperature-dependence of these values over the 25-50° C. range is preferably minor, as indicated below.

- 24 -

TABLE OF BROAD AND PREFERRED LIMITS
ON VARIATIONS IN RHEOLOGICAL PROPERTIES
(All values in 10^4 dynes/cm^2, etc.
over the 25-50° C. range)

Property	Broad Limits		Preferred Limits	
	0.25 Hz	0.01 Hz	0.25 Hz	0.01 Hz
G" (10^4 dynes/cm^2)	± 70	± 10	± 25	± 10
G' (10^4 dynes/cm^2)	± 40	± 15	± 25	± 8
Loss tangent	± 0.6	± 0.2	± 0.5	± 0.15

Actual test data indicate that an "ideal" viscoelastic PSA, within the context of this invention, would have the following properties:

	0.01 Hz	0.25 Hz
G" (dynes/cm^2)	1.1×10^5	3×10^5
G' (dynes/cm^2)	1.3×10^6	1.75×10^6
Loss tangent	0.085	0.2
J' (cm^2/dyne)	7.7×10^{-7}	6×10^{-7}
J" (cm^2/dyne)	6×10^{-8}	1×10^{-7}

With these physical properties in mind, proportions of rubbery block copolymer and end block and midblock resins can be selected to provide an effective PSA with good elastic recovery or elastic memory characteristics. The following Table of broad, preferred, and optimum proportions assumes that the rubbery block copolymer is either (1) "SOLPRENE® 418", trademark of Phillips Chemical Company for a radial isoprene-styrene block copolymer having an approximate molecular weight of 300,000, a specific gravity of 0.92, an inherent viscosity in toluene of 1.16, and an isoprene/styrene ratio of 85/15, or (2) KRATON® 1111, trademark of Shell Chemical Co. for a polystyrene-polyisoprene-polystyrene (S-I-S) block copolymer containing 21.5% styrene, the balance being isoprene. This copolymer has the previously reported tensile strength, 300% modulus, elongation at break, set at break, and Shore A hardness. The Brookfield viscosity in toluene is 1300 cps.

SOLPRENE® 418 in toluene has a viscosity of 2,900
centipoise at 25° C. Hydrogenated linear block
copolymers, including those of the
styrene-isoprene-styrene type are available according
5   to U.S. Patent No. 3,827,999; see also U.S. Patent
No. 4,089,824, which discloses the hydrogenated
butadiene analog. Such hydrogenated block copolymers
can be used in the context of this invention.
Compounds of the "KRATON® G" series (trademark of
10   Shell Chemical Company) have a saturated or
essentially saturated ethylene-butylene midblock
and, if used, are preferably used in combination
with the KRATON® 1111 or SOLPRENE® 418 type of rubbery
block copolymer.

15   The aforementioned Table of proportions is set
forth below.

TABLE OF PROPORTIONS
FOR BLOCK COPOLYMER AND RESINS

Ingredient	Amount				
	Broad	Preferred		Optimum	
	phr	wt-%	phr	wt-%	phr
Rubbery block copolymer	(100)	20-75	(100)	35-45	(100)
Midblock resin	20-150	15-50	50-125	30-50	70-120
End block resin	10-150	10-40	40-65	15-30	40-55

NOTE: phr = parts per hundred by weight, based on 100
            parts block copolymer.
       wt-%= percentage by weight of total hot melt
            PSA composition.

30   The nature of the block copolymer (linear or
branched, amount of vinyl arene units, etc.) can have
a slight effect upon the optimum formula. The best
balance of viscoelastic properties appears to be
obtained with 100 parts by weight linear,
35   high-styrene block copolymer (35-45% by weight of
the PSA), 70-78 parts by weight of the synthetic
polyterpene, and 45-55 parts by weight of a

coumarone-indene resin having a softening point in excess of 145° C. About 0.005-1 part by weight of antioxidant should be added to such a formula.

Similar formulas can provide similar results with branched block copolymers and low-styrene linear block copolymers, but it may be useful to extend the ranges up to 85 phr of polyterpene and up to 65 phr of coumarone-indene resin.

The most effective criteria for the selection of amounts and types of ingredients are believed to be (a) rheological properties such as G', G", loss tangent, J' and J", and (b) dead load creep test results. New resins and new block copolymers are constantly being discovered, and the formulation of suitable PSA's can be attempted with new materials by referring to these criteria.

It will be understood that variations in the aforementioned methods and proportions can be made without departing from the spirit and scope of this invention. For example, if the substrate to be elastic banded is heat resistant, the extruded band need not be cooled very much below its softening point and can be applied directly to the substrate while still relatively hot. However, even for such temperature resistant substrates, maintaining the extruded band at or above its softening point is neither necessary nor desirable. In another variation of the method, the extruded band need not be applied as a line or linear bead, but can be "indexed" with a moving web to provide a series of discrete circular or eliptical bands. Such "indexed" bands can provide a hat-banding effect (e.g. for mass-produced surgical caps), a gathered, banded opening for a plastic bag, a waist band, or the like. In the case of disposable diapers, however, it is not necessary for the band to form a complete circle; the essential equivalent

of a circular leg band results when the diaper is
pinned or snapped together at the child's hips.

The following non-limiting Examples illustrate
the preferred practice of this invention. In these
5   Examples, all parts and percentages are by weight
unless otherwise indicated. The following raw
materials were used in formulating the hot melt PSA's
of the Examples:

"SOLPRENE® 418": trademark for the radial
10   block copolymer described previously.

"SOLPRENE® 423": another trademark for
essentially the same radial block copolymer in a
pellet form.

"KRATON® 1111": trademark of Shell Chemical
15   Co. for the polystyrene-polyisoprene-polystyrene
copolymer described previously.

"KRATON® 1107": trademark of Shell Chemical
Co. for polystyrene-polyisoprene-polystyrene
linear block copolymer having a styrene/isoprene
20   ratio of 14/86.

"WINGTACK 95": trademark for synthetic
polyterpene resin described previously.

"KRYSTALEX® 3100": trademark of Hercules
Inc. for low molecular weight thermoplastic
25   hydrocarbon resin of the alpha-methylstyrene
type having a ring and ball softening point
of 97-103° C., an acid number less than 1.0,
a bromine number which is typically about 2,
a specific gravity at 25° C. of 1.06, and a
30   melt viscosity of 10,000 centipoise (cps) at
128° C., 1,000 cps at 152° C., and 100 cps
at 190° C. The softening point substantially
below 115° C. (typically not more than 103° C.)
indicates a spectrum of molecular weights, with
35   a significant number of resin molecules having

- 28 -

molecular weights well below those of the
relatively pure, narrow-spectrum coumarone-indene
resins which are commercially available, e.g.
as the "CUMAR" (trademark) series described
5   subsequently. (It has been found that the
higher molecular weight, higher softening point,
narrow-spectrum aromatic hydrocarbon resins are
preferred for use as "endblock" association with
the rubbery block copolymers described previously.)
10   "CUMAR LX-509": trademark of Neville
Chemical Company for coumarone-indene resin
having a softening point (by the ring and ball
technique of A.S.T.M. E-28) of at least about
155° C., a specific gravity at 25/15.6° C. of
15   1.114, and an average molecular weight (by
osmometry) of 1,120.
           "EASTMAN® Resin H-100": trademark of
·Eastman Kodak Company for a hydrocarbon resin
produced from petroleum feedstock by
20   polymerization, followed by hydrogenation.
This particular hydrocarbon resin has an acid
number less than 0.1, a density at 23° C. of
1.04 g/cm^3, a Brookfield viscosity at 190° C.
of 200 centipoise, a bromine number of 11.1,
25   and a ring and ball softening point (A.S.T.M.
E-28) which is reported to be 100° C. and in
any event is below 115°.C.
           "IRGANOX 1010": trademark of Ciba-Geigy
for an antioxidant and thermostabilizer of
30   the hindered phenol type.
           "SOLPRENE® 420": trademark for a branched,
teleblock copolymer having polystyrene terminal
blocks and a structure essentially similar to
"SOLPRENE® 423", except for a lower molecular
35   weight.

- 29 -

"KRATON® 1102": trademark for S-B-S
(styrene-butadiene-styrene) block copolymer
having a styrene/butadiene ratio of 28/72, a
Brookfield viscosity in toluene solution

5 (25 weight-%) of 1200 centipoise at 25° C., a
specific gravity of 0.94, a Shore A hardness of
62, a set at break of 10%, an elongation of 880%
(A.S.T.M. method D412 with a tensile tester jaw
separation speed of 25.4 cm/min.), a 300% modulus

10 of 281,200 Kg/m^2, and a tensile strength (same
A.S.T.M. method as the elongation determination)
of 3.23×10^6 Kg/m^2 determined on typical films
cast from a toluene solution.

"WESTON® 618": trademark of Borg Warner

15 Corporation for an antioxidant described in
U.S. Patent Nos. 3,047,608 and 3,205,269, i.e.
an antioxidant which is reported to be di(stearyl)
pentaerythritol diphosphite.

For convenience of sample preparation, the

20 antioxidants and pigments were sometimes omitted
from the exemplary formulations which follow. Since
incompatibility between resins and various portions
of the rubbery block copolymer tend to be minimized
in the molten state, the order of addition of

25 ingredients is not usually critical. It is generally
preferred to begin with one of the relatively larger
components such as the rubbery block copolymer and add
the tackifiers and other resins to it, e.g. adding
the synthetic terpene "midblock" resin next, followed

30 by the "endblock" resin. Samples can be prepared by
blending in a solvent medium; however, the data
obtained from such samples is believed to be less
reliable as compared to samples formulated in the
molten state. Samples can be solvent-cast to films

35 ranging from 100 to 200 micrometers for test purposes,

even though the industrial practice of this invention
involves extrusion of the hot melt PSA.

<div align="center">Comparative Example A</div>

The following ingredients were blended in a
5 heated mixer in the indicated amounts.

Wt.-%	Ingredient
65.4	Radial isoprene-styrene elastomeric block copolymer (SOLPRENE® 423).
32.7	High softening-point, high molecular weight coumarone-indene resin (CUMAR® LX-509).
0.2	Phosphite ester antioxidant (WESTON® 618).
0.2	Hindered phenol antioxidant ("IRGANOX" 1010).
1.5	Titanium dioxide pigment (rutile, alumina-treated).

A purpose of this Example was to evaluate the effect
upon rheology and room-temperature 180° peel strength
20 when the radial block copolymer/resin blend was
provided with large crystalline (vinyl arene) domains
and minimally tackified or plasticized rubbery
(elastomeric) domains,. in this case no "midblock"
resin. According to the scientific and patent
25 literature, the coumarone-indene resin probably
became associated with the polystyrene end blocks of
the radial elastomeric block copolymer.

<div align="center">Comparative Example B</div>

The following formula is a rubbery block
30 copolymer/tackifier resin blend which would have
low tack and peel and would have significant elastomeric
behavior at normal ambient temperatures. The formula
theoretically contains no "endblock" resin.

— 31 —

Wt.-%	Ingredient
50	Linear S-B-S (polystyrene-polybutadiene-polystyrene) elastomeric block copolymer (KRATON® 1102).
35	Hydrocarbon resin (EASTMAN® Resin H-100).
15	Staybelite Ester 10 (HERCULES® glycerol ester of hydrogenated rosin, 83° C. softening point).

<u>Comparative Example C</u>

This formula was similar to that of Example B, except that an S-I-(polystyrene-polyisoprene-) radial block copolymer was blended with a different tackifier.

Wt.-%	Ingredient
50	Radial S-I- elastomeric block copolymer (SOLPRENE® 418).
50	Polyterpene resin (WINGTACK® 95).

<u>Comparative Example D</u>

This formula had both "endblock" resin and "midblock" resin in addition to the S-I-S block copolymer; however, more than 80% by weight of the "endblock" resin was a relatively low molecular weight, low softening point material. The block copolymer was also a relatively lower molecular weight material. Room temperature performance of the formula would be expected to include poor PSA properties and some elastomeric behavior, but at higher temperatures (e.g. 37° C.), performance would be unpredictable.

- 32 -

Wt.-%	Ingredient
40.0	Low molecular weight, radial, elastomeric block-copolymer (SOLPRENE® 420).
0.1	Phosphite ester antioxidant (WESTON® 618).
0.1	Hindered phenol antioxidant ("IRGANOX" 1010).
1.0	Titanium dioxide pigment)rutile, alumina-treated).
32.8	Hydrocarbon resin (EASTMAN® Resin H-100).
4.0	High molecular weight coumarone-indene (CUMAR® LX-509).
22.0	Alpha-methylstyrene resin (typical softening point: 100° C.)("KRYSTALEX" 3100 [trademark]).

Comparative Example E

This formula appeared to conform to all of the criteria of a formula of this invention, except that the amount of "endblock" resin was relatively high and the amount of polyterpene tackifier was relatively low.

Wt.-%	Ingredient
45.0	High molecular weight, radial S-I- elastomeric block copolymer (SOLPRENE® 418).
15.0	Polyterpene resin (WINGTACK® 95).
40.0	High molecular weight, high softening point coumarone-indene (CUMAR® LX-509).

Examples 1 - 5

The formulas for Examples 1 through 5 are set forth in the following Table.

INGREDIENTS IN WEIGHT–%

Example	S-I-S Linear Block Copolymer	S-I Radial Block Copolymer	Polyterpene Resin (WINGTACK® 95)	Coumarone-indene (CUMAR® LX-509)	Miscella-neous (1)
1	43.6[2]	--	32.7	21.8	1.9
2	43.6[3]	--	32.7	21.8	1.9
3	--	43.6[4]	32.7	21.8	1.9
4	39.6[3]	--	32.4	26.1	1.9
5	--	38.0[5]	45.0	17.0	--

NOTES

(1) This "miscellaneous" component consisted of 0.2% WESTON® 618, 0.2% "IRGANOX" 1010, and 1.5% titanium dioxide pigment (rutile, alumina-treated).

(2) KRATON® 1111, described previously.

(3) KRATON® 1107, described previously.

(4) SOLPRENE® 423, described previously.

(5) SOLPRENE® 418, described previously.

- 34 -

Rheological Testing

The G", G', loss tangent (G"/G'), J", and J'
were determined, as described previously, at 25° and
50° C. and at 0.01 and 0.25 Hz. The results for
5 the Comparative Examples and Examples 1-5 are set
forth below. All G" and G' data are in 10^4
dynes/cm^2, and all J" and J' data are in 10^{-8}
cm^2/dyne. For purposes of comparison, typical data
for natural rubber (at 0.25 Hz) are included.

TABLE I: Rheological Testing
Frequency: 0.01 Hz
Temperature: 25° C. and 50° C.

Example	G" 25°	G" 50°	G' 25°	G' 50°	loss tan 25°	loss tan 50°	J" 25°	J" 50°	J' 25°	J' 50°
A	121	140	89.5	323	1.351	.428	53.4	11.1	39.5	26.2
B	16.6	21.7	129	74.5	.129	.290	9.86	35.7	76.3	124
C	4.65	6.16	67.4	71.7	.069	.086	10.2	11.9	148	138
D	53.3	22.7	173	65.3	.308	.408	16.3	62.9	52.8	131
E	20.2	21.9	345	336	.058	.065	1.66	1.91	28.9	29.6
1	12.8	9.28	128	131	.100	.071	7.74	5.40	77.4	76.0
2	13.6	12.4	146	151	.093	.082	6.31	5.39	67.9	65.8
3	14.2	15.6	161	163	.088	.096	5.41	5.85	61.6	60.8
4	11.4	10.1	130	123	.087	.082	6.59	6.61	76.3	80.8
5	13.3	5.23	69.7	64.9	.191	.081	26.5	12.3	138	153

TABLE II: Rheological Testing
Frequency: 0.25 Hz
Temperature: 25° C. and 50° C.

Example	G"		G'		loss tan		J"		J'	
	25°	50°	25°	50°	25°	50°	25°	50°	25°	50°
A	180	173	148	323	1.215	.440	33.1	9.38	27.3	21.3
B	31.5	19.3	171	110	.187	.173	10.7	15.1	56.5	88.3
C	15.0	5.48	78.9	82.3	.190	.067	23.2	8.16	122	121
D	138	45.7	311	130	.442	.351	11.8	24.0	26.9	68.5
E	34.3	23.0	386	372	.089	.062	2.29	1.66	25.7	26.8
1	33.1	15.5	164	152	.203	.102	12.0	6.64	58.6	65.1
2	25.7	15.6	175	176	.147	.089	8.23	5.03	55.9	56.9
3	32.1	18.1	197	190	.163	.096	8.06	10.3	49.4	52.2
4	31.3	15.1	167	145	.188	.104	10.9	7.09	57.8	68.2
5	73.3	9.68	115	76.4	.639	.127	39.6	70.2	61.7	129
Natural Rubber (estimated)	40	40	800	800	.05	.05	--	--	--	--

For Example A, the high G' values and the
temperature dependency of these values indicate
unsuitability, according to the principles of this
invention. The high G' may, it is believed, indicate

5   good elastic behavior, but, conversely, poor PSA
performance. The rheological parameters were
confirmed by 180° peel data (PSTC-1). Even 24 hours
after formation of the PSA bond, no PSTC-1 value could
be obtained. "Dead load deformation" values were

10 acceptable, ranging from 0% to only 6% throughout the
25-50° C. range.

For Example B, the temperature dependence of G'
also indicates unsuitability, which was reflected
in the "dead load deformation" data. These data

15 were as follows: 20% at room temperature, 344% at
37.8° C., and 528% at 43.3° C. Cohesive failure
occurred at 48.9° C.

For Example C, G" at .01 Hz (25° C.) was
marginal, and at .25 Hz (50° C.) was very low; G'

20 data at .01 Hz were marginal. This analysis of the
rheological parameters was confirmed by "dead load
deformation" data: 6% at room temperature, 44% at
37.8° C., 92% at 43.3° C., and cohesive failure at
48.9° C. The PSTC-1 values after 24 hours were

25 acceptable (2770 g/25.4 mm-width), indicating the
ability to wet out a substrate (but inadequate
resistance to heat set).

Example D had poor PSA behavior, and
performed poorly in the "dead load deformation"

30 test. It is believed that the high G" and, most
important, temperature dependence of G' were
significant in these regards.

The high coumarone-indene content of Example E
was believed to be reflected in the high G' values.

35 The PSA behavior of Example E was marginal (e.g.

- 38 -

910 g/25.4 mm-width in PSTC-1), but a sample of
Example E performed adequately in the "dead load
deformation" test.

5   Examples 1-4 performed well in terms of both
(1) PSTC-1 (180° peel) values taken at room temperature
after 24 hours PSA bond formation and (2) "dead
load deformation". Example 5 showed excellent
PSTC-1 performance but nearly failed the "dead
load deformation" test. The 24-hour PSTC-1 values
10 for Examples 1, 3, 4, and 5 are set forth below.
All samples tested were 25.4 mm in width, and the
values are reported in grams per 25.4 mm-width.

        Example 1:   2670 g/25.4 mm

        Example 3:   2170 g/25.4 mm

15   Example 4:   2130 g/25.4 mm

        Example 5:   3580 g/25.4 mm*

    *determined immediately rather than after 24 hours.
PSTC-1 values above 4500 g/25.4 mm or even 4000
g/25.4 mm are difficult to obtain in practice without
20 sacrificing other desired properties.

Dead Load Deformation Test

     All of the products tested were manufactured
using a high shear, double arm mixer heated to 200°
C. Inert gas was used throughout processing to
25 minimize degradation.

     Samples taken from each batch were pressed between
release paper using a Carver laboratory press. The
release paper was heated in a forced air oven to
200° C. for 15 minutes to drive out residual moisture.
30 The press was equipped with heated jaws set at
200° C. and was shimmed to yield an adhesive thickness
of 50 micrometers.

     A dwell time of approximately 5 seconds at
3.5×10^6 Kg/m^2 was generally sufficient to form
35 an air-free film.

– 39 –

Dumbbell-shaped specimens were cut from the essentially air-free films using a standard striking die.

Marks were placed on the reduced section of a
5 specimen approximately equidistant from its center and perpendicular to its longitudinal axis. The centers of the marks were 25.0 mm ± 0.5 mm apart.

The specimens were fastened in a vertical position at test temperature and weights equalling 1500
10 gms/cm^2 of cross sectional area were attached. The specimens were conditioned in this mode for a period of 3 hours. After 3 hours, the weights were removed and the specimens allowed to retract and equilibrate at 25° C. for 5 minutes. The distance between marks
15 was remeasured and the percent "dead load creep" or elongation (permanent longitudinal deformation or set) was calculated as follows:

Elongation percent $= L_2 - L_1/L_1 \times 100$

wherein L_2 = measured distance between marks on the
20 conditioned specimen, and L_1 = original distance between marks.

This procedure is a modification of A.S.T.M. D-412 (tension testing of vulcanized rubber) and the dumbbell-shaped samples were formed using Die C.
25 The dead load deformation data for Examples 1, 3, 4, and 5 are set forth below.

TABLE III

PERMANENT DEFORMATION FROM 1500 g/cm^2

AT VARIOUS TEMPERATURES

Example	Room Temperature	37.8° C.	43.3° C.	48.9° C.
1	2%	4%	8%	8%
3	2%	10%	30%	30%
4	2%	10%	24%	36%
5	6%	34%	56%	74%

- 40 -

In the case of Example 5, some temperature dependency of G", G', and loss tangent should be noted at 0.25 Hz, and the deformation data are believed to reflect a similar temperature dependency.
5　Furthermore, G' at 0.01 Hz was considered marginal for this formula.

The data for Examples 1-5 are believed to establish that a variety of relatively high molecular weight, relatively high vinyl-arene elastomeric
10　block copolymers can be used in this invention, e.g. both the radial and linear types. Blends of these various types of block copolymers (e.g. 1/99 - 99/1) will provide suitable elastomeric bases for admixture with endblock and midblock resins or
15　other PSA-forming materials. Examples 1-5 are also believed to demonstrate the value of a good balance between high softening point "endblock" resin and "midblock" resin in the preferred compositions.

Samples of Examples 1-5 were tested for tensile
20　strength at 500% elongation at room temperature (20-25° C.). It was found that the tensile strength exceeded 35,000 Kg/m^2 for all samples. For Examples 1-4, this measurement was significantly higher, generally in excess of 70,000 Kg/m^2, indicating
25　good cohesiveness.

To provide a further standard of comparison for the rheological data obtained from Examples 1-5, data regarding untackified, unextended, linear A-B-A block copolymers (e.g. of the KRATON® type)
30　were obtained for 0.25 Hz/room temperature conditions. These data indicate that G' is typically above 300 x 10^4 dynes/cm^2 and G" is typically above 50 x 10^4 dynes/cm^2 and even, in some cases, above 100 x 10^4 dynes/cm^2. Like vulcanized natural rubber, these
35　block copolymers exhibit excellent elastomeric behavior but, in the absence of tackifying resins, essentially no PSA behavior.

This application is a continuation-in-part of our
co-pending application, U.S. Serial No. 966,794, filed
December 6, 1978.

WE CLAIM: - 42 -

    1.   A method for imparting elongation-resistant gathers to portions of a generally non-elastomeric film, membrane, or web substrate, comprising the steps of:

    (a)   extruding a band comprising a viscoelastic hot melt pressure-sensitive adhesive, said hot melt pressure-sensitive adhesive comprising a rubbery block copolymer which includes a rubbery midblock portion and which is terminated with crystalline vinyl arene end blocks, said hot melt pressure-sensitive adhesive having a ball and ring softening point within the range of 65° to 240° C., being elastomeric above its glass transition temperature and at temperatures up to at least 37° C., and having the following rheological properties at 0.01-0.25 Hz and throughout the range of 25-50° C.:

    loss modulus: 5×10^4 to 100×10^4 dynes/cm^2

    storage modulus: 65×10^4 to 225×10^4 dynes/cm^2

    loss tangent: 0.03 to 1.0;

    (b)   cooling the thus-extruded band to a temperature below its softening point but above its glass transition temperature;

    (c)   bringing the thus-cooled band into contact with a surface of a moveable, flexible, generally non-elastomeric continuous film, membrane, or web substrate having a thickness less than about 3 millimeters and forming a pressure-sensitive adhesive bond generally by means of pressure, whereby the thus-banded substrate can have elongation-resistant gathers at least along the area of the pressure-sensitive adhesive bond between said band and said substrate;

(d) cutting the resulting banded substrate
into portions suitable for use as elastic-banded
articles, said articles having elongation-resistant
gathers at least along said area of the
pressure-sensitive adhesive bond, due to the
expandability of said gathers and the
elastomeric properties of said band.

2. A method according to claim 1 in which said
moveable flexible continuous film, membrane, or web
substrate is kept continuously moving during said
steps (a), (b), and (c).

3. A method according to claim 1 wherein said
steps (b) and (c) are carried out simultaneously.

4. A method according to claim 3 wherein gathers
are introduced into said banded substrate by placing
said band under tension subsequent to said step (a)
and prior to said step (d), whereby said substrate is
wrinkled into gathers through contraction of the
tensioned band.

5. A method according to claim 1, wherein said
substrate is a polymeric film less than 0.5 mm thick
or a woven or nonwoven web, and wherein said
elastic-banded articles of step (d) are garments or
body-encircling members.

6. A method according to claim 1 wherein said
hot melt pressure-sensitive adhesive comprises:

(a) said rubbery block copolymer;

(b) 20-150 parts by weight, per 100 parts
by weight of said rubbery block copolymer, of
a tackifying resin generally compatible with and
generally associated with said midblock portion;
and

(c) 10-150 parts by weight, per 100 parts
by weight of said rubbery block copolymer, of
an aromatic, essentially hydrocarbon resin having

– 44 –

a glass transition temperature and a softening point above those of the said end blocks and said tackifying resin; said aromatic, essentially hydrocarbon resin being generally compatible with and generally associated with said end blocks.

7. A method according to claim 1 wherein the said loss modulus, storage modulus, and loss tangent remain generally constant throughout the temperature range of 25-50° C. with respect to properties determined at both 0.25 and 0.01 Hz, at least within the following limits:

	0.25 Hertz	0.01 Hertz
loss modulus	$\pm\ 70 \times 10^4$ dynes/cm^2	$\pm\ 10 \times 10^4$ dynes/cm^2
storage modulus	$\pm\ 40 \times 10^4$ dynes/cm^2	$\pm\ 15 \times 10^4$ dynes/cm^2
loss tangent	$\pm\ 0.6$	$\pm\ 0.2.$

8. A method according to claim 7 wherein said pressure-sensitive adhesive has the following rheological properties throughout the temperature range of 25-50° C.:

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	5-25	10-100
storage modulus, 10^4 dynes/cm^2	75-200	75-200
loss tangent	0.05-0.3	0.08-1.0
storage compliance, 10^{-8} cm^2/dyne	40-100	40-100
loss compliance, 10^{-8} cm^2/dyne	2-20	5-40.

9. A method according to claim 8 wherein said rheological properties are as follows:

- 45 -

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	8-16	15-35
storage modulus, 10^4 dynes/cm^2	100-175	120-200
loss tangent	0.07-0.10	0.1-0.25
storage compliance, 10^{-8} cm^2/dyne	60-80	50-70
loss compliance, 10^{-8} cm^2/dyne	5-8	5-15.

10. A method according to claim 1 wherein the
dead load deformation of said pressure-sensitive
adhesive, tested at room temperature, 37.8° C.,
43.3° C., and 48.9° C. for 3 hours under 1500g/cm^2,
is less than 50%, where dead load deformation = the
increased length minus the original length divided
by the original length of a sample at least 25 mm
in length.

11. A method for imparting elongation-resistant
gathers to portions of a generally non-elastomeric
substrate by means of a pressure-sensitive adhesive
tape, comprising the steps of:

(a) unreeling a length of pressure-sensitive
adhesive tape from a convoluted roll of hot melt
pressure-sensitive adhesive tape comprising a hot
melt pressure-sensitive adhesive layer adhered
to a flexible continuous backing, said roll
being maintained at a temperature below the
softening point of said hot melt pressure-sensitive
adhesive layer; said hot melt pressure-sensitive
adhesive layer being an elongated extrudate at
least 50 microns in thickness comprising (i) a
rubbery block copolymer which includes a rubbery
midblock portion and which is terminated with
crystalline vinyl arene blocks; (ii) 20-150 parts
by weight, per 100 parts by weight of said

rubbery block copolymer, of a tackifying resin
generally compatible with and generally associated
with said midblock portion; and (iii) 10-150
parts by weight, per 100 parts by weight of

5   said rubbery block copolymer, of an aromatic,
essentially hydrocarbon resin having a glass
transition temperature and a softening point
above those of the said end blocks and said
tackifying resin; said aromatic, essentially

10 hydrocarbon resin being generally compatible
and generally associated with said end blocks;
said hot melt adhesive having a ball and ring
softening point within the range of 65° to 240°
C. and being elastomeric above its glass transition

15 temperature and at temperatures up to at least
37° C.;

        (b)   placing said length of the
pressure-sensitive adhesive layer of said tape
under tension; and

20   (c)   bringing an exposed surface of the
pressure-sensitive adhesive layer into contact
with said substrate and forming a
pressure-sensitive adhesive band generally by
means of pressure; whereby the thus-banded

25 substrate can have elongation-resistant gathers
at least along the area of the pressure-sensitive
adhesive bond between said band and said
substrate.

        12.   A method according to claim 11, wherein the

30 backing of said pressure-sensitive adhesive tape is a
generally non-elastomeric release liner which is
delaminated from said tape prior to said step (c).

        13.   A method according to claim 11, wherein
the backing of said pressure-sensitive adhesive tape

35 is elastomeric.

14. A method according to claim 13 wherein said
tape has been formed by coextruding said backing
and said pressure-sensitive adhesive layer, and
wherein the exposed surface of said backing has been
5 provided with release characteristics.

15. A viscoelastic hot melt pressure-sensitive
adhesive consisting essentially of:

   (a) 20-75% by weight of a rubbery block
copolymer which comprises a rubbery polyisoprene
10 midblock portion and a plurality of end blocks
comprising   . a crystalline poly(vinylarene);

   (b) 50-125 parts by weight, per 100 parts
   _ by weight of said block copolymer, of a terpene
tackifying resin generally compatible with and ·
15 generally associated with said midblock portion;

   (c) 40-65 parts by weight, per 100 parts by
weight of said radial block copolymer, of a
coumarone-indene resin having a glass transition
temperature·. and a softening point above
20 about 115° C.;

      the proportions of said components (a), (b),
and (c) being selected to provide the following
pressure-sensitive adhesive and rheological
properties:

25   (i) a tensile strength at 500%
          elongation, determined at 20-25° C., of
          at least 50 pounds per square inch;

             (ii) a 180° peel resistance, according
          to PSTC-1, determined at 20-25° C. 24 hours
30   after formation of the pressure-sensitive
          adhesive bond, of at least about 450 grams
          per 25.4 mm-width;

             (iii) a dead load deformation, tested at
          room temperature, 37.8° C., 43.3° C.,
35   and 48.9° C. for 3 hours under 1500 g/cm^2,

is less than 50%, where dead load
. deformation = the increased length minus
the original length divided by the
.original length of a sample at least 25 mm
5   in length;

      (iv) the following loss modulus, storage
modulus, and loss tangent values at 0.01-0.25
Hz throughout the temperature range of
25-50° C.:

10   loss modulus: 5×10^4 to 100×10^4
                                   dynes/cm^2

           storage modulus: 65×10^4 to 225×10^4
                                   dynes/cm^2

           loss tangent: 0.03 to 1.0.

15   16. A viscoelastic hot melt pressure-sensitive
adhesive according to claim 15, wherein said adhesive
consists essentially of:

      35-55% by weight of said block copolymer;

      25-45% by weight of said terpene tackifying
20 resin, said tackifying resin having been
synthetically derived from a mixed olefin
feedstock;

      15-30% by weight of said coumarone-indene
resin, said coumarone-indene resin having a
25 softening point of at least about 140° C.;

      0-5% by weight of substantially inert
extenders, fillers, pigments, and antioxidants;
the foregoing proportions being selected to provide
the following results for the properties defined in
30 said paragraph (iv):

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	5-25	10-100
storage modulus, 10^4 dynes/cm^2	75-200	75-200
loss tangent	0.05-0.3	0.08-1.0
storage compliance, 10^{-8} cm^2/dyne	40-100	40-100
loss compliance, 10^{-8} cm^2/dyne	2-20	5-40.

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1. A method for imparting elongation-resistant gathers to portions of a generally non-elastomeric film, membrane, or web substrate, comprising the steps of:

(a) extruding a band comprising a viscoelastic hot melt pressure-sensitive adhesive, said hot melt pressure-sensitive adhesive comprising a rubbery block copolymer which includes a rubbery midblock portion and which is terminated with crystalline vinyl arene end blocks, said hot melt pressure-sensitive adhesive having a ball and ring softening point within the range of 65° to 240° C., being elastomeric above its glass transition temperature and at temperatures up to at least 37° C., and having the following rheological properties at 0.01-0.25 Hz and throughout the range of 25-50° C.:

loss modulus: 5×10^4 to 100×10^4 dynes/cm^2

storage modulus: 65×10^4 to 225×10^4 dynes/cm^2

loss tangent: 0.03 to 1.0;

(b) cooling the thus-extruded band to a temperature below its softening point but above its glass transition temperature;

(c) bringing the thus-cooled band into contact with a surface of a moveable, flexible, generally non-elastomeric continuous film, membrane, or web substrate having a thickness less than about 3 millimeters and forming a pressure-sensitive adhesive bond generally by means of pressure, whereby the thus-banded substrate can have elongation-resistant gathers at least along the area of the pressure-sensitive adhesive bond between said band and said substrate;

(d)   cutting the resulting banded substrate
into portions suitable for use as elastic-banded
articles, said articles having elongation-resistant
gathers at least along said area of the
pressure-sensitive adhesive bond, due to the
expandability of said gathers and the
elastomeric properties of said band.

   2.   A method according to claim 1 in which said
moveable flexible continuous film, membrane, or web
substrate is kept continuously moving during said
steps (a), (b), and (c).

   3.   A method according to claim 1 wherein said
steps (b) and (c) are carried out simultaneously.

   4.   A method according to claim 3 wherein gathers
are introduced into said banded substrate by placing
said band under tension subsequent to said step (a)
and prior to said step (d), whereby said substrate is
wrinkled into gathers through contraction of the
tensioned band.

   5.   A method according to claim 1, wherein said
substrate is a polymeric film less than 0.5 mm thick
or a woven or nonwoven web, and wherein said
elastic-banded articles of step (d) are garments or
body-encircling members.

   6.   A method according to claim 1 wherein said
hot melt pressure-sensitive adhesive comprises:
      (a)   said rubbery block copolymer;
      (b)   20-150 parts by weight, per 100 parts
   by weight of said rubbery block copolymer, of
   a tackifying resin generally compatible with and
   generally associated with said midblock portion;
   and
      (c)   10-150 parts by weight, per 100 parts
   by weight of said rubbery block copolymer, of
   an aromatic, essentially hydrocarbon resin having

a glass transition temperature and a softening point above those of the said end blocks and said tackifying resin; said aromatic, essentially hydrocarbon resin being generally compatible with and generally associated with said end blocks.

7. A method according to claim 1 wherein the said loss modulus, storage modulus, and loss tangent remain generally constant throughout the temperature range of 25-50° C. with respect to properties determined at both 0.25 and 0.01 Hz, at least within the following limits:

	0.25 Hertz	0.01 Hertz
loss modulus	$\pm\ 70 \times 10^4$ dynes/cm^2	$\pm\ 10 \times 10^4$ dynes/cm^2
storage modulus	$\pm\ 40 \times 10^4$ dynes/cm^2	$\pm\ 15 \times 10^4$ dynes/cm^2
loss tangent	$\pm\ 0.6$	$\pm\ 0.2.$

8. A method according to claim 7 wherein said pressure-sensitive adhesive has the following rheological properties throughout the temperature range of 25-50° C.:

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	5-25	10-100
storage modulus, 10^4 dynes/cm^2	75-200	75-200
loss tangent	0.05-0.3	0.08-1.0
storage compliance, 10^{-8} cm^2/dyne	40-100	40-100
loss compliance, 10^{-8} cm^2/dyne	2-20	5-40.

9. A method according to claim 8 wherein said rheological properties are as follows:

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	8–16	15–35
storage modulus, 10^4 dynes/cm^2	100–175	120–200
loss tangent	0.07–0.10	0.1–0.25
storage compliance, 10^{-8} cm^2/dyne	60–80	50–70
loss compliance, 10^{-8} cm^2/dyne	5–8	5–15.

10. A method according to claim 1 wherein the dead load deformation of said pressure-sensitive adhesive, tested at room temperature, 37.8° C., 43.3° C., and 48.9° C. for 3 hours under 1500g/cm^2, is less than 50%, where dead load deformation = the increased length minus the original length divided by the original length of a sample at least 25 mm in length.

11. A method for imparting elongation-resistant gathers to portions of a generally non-elastomeric substrate by means of a pressure-sensitive adhesive tape, comprising the steps of:

(a) unreeling a length of pressure-sensitive adhesive tape from a convoluted roll of hot melt pressure-sensitive adhesive tape comprising a hot melt pressure-sensitive adhesive layer adhered to a flexible continuous backing, said roll being maintained at a temperature below the softening point of said hot melt pressure-sensitive adhesive layer; said hot melt pressure-sensitive adhesive layer being an elongated extrudate at least 50 microns in thickness comprising (i) a rubbery block copolymer which includes a rubbery midblock portion and which is terminated with crystalline vinyl arene blocks; (ii) 20-150 parts by weight, per 100 parts by weight of said

54

rubbery block copolymer, of a tackifying resin
generally compatible with and generally associated
with said midblock portion; and (iii) 10-150
parts by weight, per 100 parts by weight of
said rubbery block copolymer, of an aromatic,
essentially hydrocarbon resin having a glass
transition temperature and a softening point
above those of the said end blocks and said
tackifying resin; said aromatic, essentially
hydrocarbon resin being generally compatible
and generally associated with said end blocks;
said hot melt adhesive having a ball and ring
softening point within the range of 65° to 240°
C. and being elastomeric above its glass transition
temperature and at temperatures up to at least
37° C.;

    (b) placing said length of the
pressure-sensitive adhesive layer of said tape
under tension; and

    (c) bringing an exposed surface of the
pressure-sensitive adhesive layer into contact
with said substrate and forming a
pressure-sensitive adhesive band generally by
means of pressure; whereby the thus-banded
substrate can have elongation-resistant gathers
at least along the area of the pressure-sensitive
adhesive bond between said band and said
substrate.

    12. A method according to claim 11, wherein the
backing of said pressure-sensitive adhesive tape is a
generally non-elastomeric release liner which is
delaminated from said tape prior to said step (c).

    13. A method according to claim 11, wherein
the backing of said pressure-sensitive adhesive tape
is elastomeric.

14. A method according to claim 13 wherein said tape has been formed by coextruding said backing and said pressure-sensitive adhesive layer, and wherein the exposed surface of said backing has been provided with release characteristics.

15. A viscoelastic hot melt pressure-sensitive adhesive consisting essentially of:

(a) 20-75% by weight of a rubbery block copolymer which comprises a rubbery polyisoprene midblock portion and a plurality of end blocks comprising a crystalline poly(vinylarene);

(b) 50-125 parts by weight, per 100 parts by weight of said block copolymer, of a terpene tackifying resin generally compatible with and generally associated with said midblock portion;

(c) 40-65 parts by weight, per 100 parts by weight of said radial block copolymer, of an aromatic, essentially hydrocarbon resin having a glass transition temperature and a softening point above about 115° C.;

the proportions of said components (a), (b), and (c) being selected to provide the following pressure-sensitive adhesive and rheological properties:

(i) a tensile strength at 500% elongation, determined at 20-25° C., of at least 50 pounds per square inch;

(ii) a 180° peel resistance, according to PSTC-1, determined at 20-25° C. 24 hours after formation of the pressure-sensitive adhesive bond, of at least about 450 grams per 25.4 mm-width;

(iii) a dead load deformation, tested at room temperature, 37.8° C., 43.3° C., and 48.9° C. for 3 hours under 1500 g/cm^2,

is less than 50%, where dead load deformation = the increased length minus the original length divided by the original length of a sample at least 25 mm in length;

       (iv)   the following loss modulus, storage modulus, and loss tangent values at 0.01-0.25 Hz throughout the temperature range of 25-50° C.:

        loss modulus: 5×10^4 to 100×10^4 dynes/cm^2

        storage modulus: 65×10^4 to 225×10^4 dynes/cm^2

        loss tangent: 0.03 to 1.0.

    16.   A viscoelastic hot melt pressure-sensitive adhesive according to claim 15, wherein said adhesive consists essentially of:

       35-55% by weight of said block copolymer;

       25-45% by weight of said terpene tackifying resin, said tackifying resin having been synthetically derived from a mixed olefin feedstock;

       15-30% by weight of said aromatic, essentially hydrocarbon resin, said aromatic, essentially hydrocarbon resin having a softening point of at least about 140° C.;

       0.5% by weight of substantially inert extenders, fillers, pigments, and antioxidants; the foregoing proportions being selected to provide the following results for the properties defined in said paragraph (iv):

17.   A viscoelastic hot melt pressure-sensitive
adhesive consisting essentially of:
      (a)   about 35 to about 55% by weight of a
rubbery block copolymer which comprises a rubbery
polyisoprene midblock portion and a plurality of
end blocks comprising a crystalline poly(vinylarene);
      (b)   about 25 to 45% by weight of a terpene
tackifying resin generally compatible with and
generally associated with said midblock portion;
      (c)   about 10 to about 30% by weight of an
aromatic, essentially hydrocarbon resin having a
glass transition temperature and a softening point
above about 115° C., said essentially hydrocarbon
resin being generally compatible with said end
blocks of said rubbery block copolymer;
      the proportions of said components (a), (b),
and (c) being selected to provide the following
pressure-sensitive adhesive and rheological
properties:
         (i)   a tensile strength at 500% elongation,
    determined at 20-25° C., of at least 50 pounds
    per square inch;
         (ii)   a 180° peel resistance, according
    to PSTC-1, determined at 20-25° C. 24 hours
    after formation of the pressure-sensitive
    adhesive bond, of at least about 1500 grams
    per 25.4 mm-width;
         (iii)   a dead load deformation, tested at
    room temperature, 37.8° C., 43.3° C., and
    48.9° C. for 3 hours under 1500 g/cm^2, of less
    than 100%, where dead load deformation = the
    increased length minus the original length
    divided by the original length of a sample at
    least 25 mm in length;
         (iv)   the following loss modulus, storage
    modulus, and loss tangent values at 0.01 and 0.25
    Hz throughout the temperature range of 25-50°
    C.:

	0.01 Hz	0.25 Hz
loss modulus, 10^4 dynes/cm^2	5-100	0-100
storage modulus, 10^4 dynes/cm^2	65-225	75-200
loss tangent	0.03-1.0	0.08-1.0

wherein the said values remain generally within the following limits, throughout said temperature range, at 0.25 Hz:

loss modulus $\pm$ 70 x 10^4 dynes/cm^2

storage modulus $\pm$ 40 x 10^4 dynes/cm^2

loss tangent $\pm$ 0.6.

18. A viscoelastic hot melt pressure-sensitive adhesive according to claim 17, wherein said adhesive consists essentially of:

up to 85 parts by weight, per 100 parts by weight of said block copolymer, of said terpene tackifying resin, said tackifying resin having been synthetically derived from a mixed olefin feedstock;

up to 65 parts by weight, per 100 parts by weight of said block copolymer, of said essentially hydrocarbon resin, said essentially hydrocarbon resin having a softening point of at least about 140° C.;

0-5% by weight of substantially inert extenders, fillers, pigments, and antioxidants;

the foregoing proportions being selected to provide the following loss modulus, storage modulus, loss tangent, storage compliance, and loss compliance values, throughout the temperature range of 25-50° C., at 0.01 Hz:

loss modulus, 10^4 dynes/cm^2	5-25
storage modulus, 10^4 dynes/cm^2	75-200
loss tangent	0.05-0.3
storage compliance, 10^{-8} cm^2/dyne	40-100
loss compliance, 10^{-8} cm^2/dyne	2-20

and the following storage compliance and loss compliance values, throughout said temperature range at 0.25 Hz:

storage compliance, 10^{-8} cm^2/dyne	40-100
loss compliance, 10^{-8} cm^2/dyne	5-40.

19. A viscoelastic hot melt pressure sensitive
adhesive according to claim 18, wherein said aromatic,
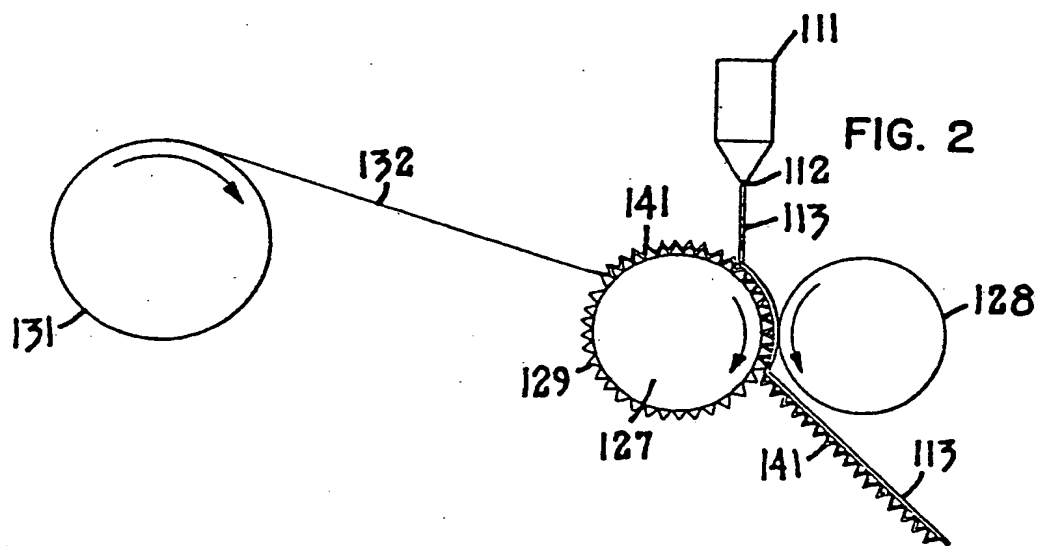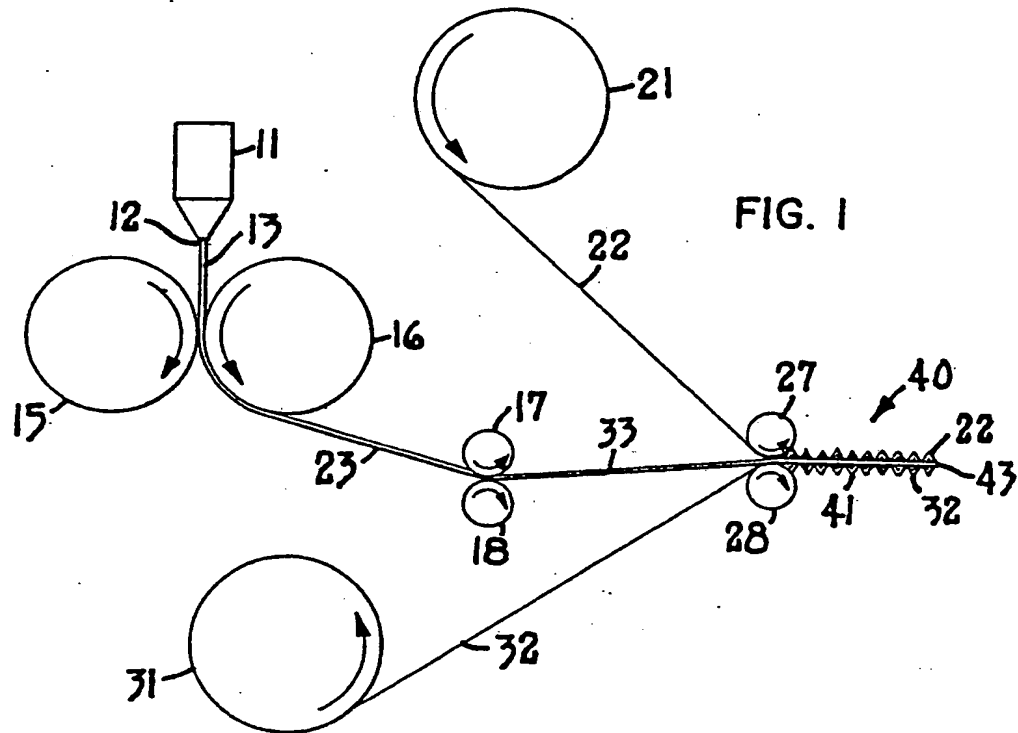essentially hydrocarbon resin is a coumarone-indene
resin.

STATEMENT UNDER ARTICLE 19

The changes in claims 15 and 16 are based upon original claim 6, paragraph (c); see the paragraph bridging pages 44 and 45 of the application. New claims 17-19 are based upon original claims 15 and 16 and also original claims 6 (paragraph [c]) and the last six lines of claim 7. New language and numerical limits in paragraph (c) of new claim 17 are based upon original claim 16, original claim 6, paragraph (c), and page 25, line 25 (page 26 in the original page numbering) of the instant specification. The PSTC-1 value in claim 17, paragraph (c) (ii) is based upon page 22, line 34 (originally numbered page 23) of the instant specification. The "dead load deformation" value of "less than 100%" is based upon page 23, line 8 (page 24 in the original numbering) of the instant specification. Parts per hundred (phr) limits for the terpene resin and the essentially hydrocarbon resin in claim 18 are based upon page 26, lines 4-8 (originally numbered page 27) of the instant specification.

FIG. 1

FIG. 2

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ³

According to International Patent Classification (IPC) or to both National Classification and IPC

INT. Cl. B31 F1/12, 31/20; B32B 31/08; C08K5/01; A41D 9/14
U.S. Cl. 156/84, 183, 205, 210, 244.19, 244.27, 269; 260/29.8
525/96, 98;

II. FIELDS SEARCHED

Classification System	Minimum Documentation Searched ⁴
	Classification Symbols
U. S.	156/84, 205, 210, 244.19, 244.27, 269, 260/29.8
	525/96, 98; 2/401

Documentation Searched other than Minimum Documentation
to the Extent that such Documents are Included in the Fields Searched ⁵

None

III. DOCUMENTS CONSIDERED TO BE RELEVANT ¹⁴

Category *	Citation of Document, ¹⁶ with indication, where appropriate, of the relevant passages ¹⁷	Relevant to Claim No. ¹⁸
A	US, A, 3,917,607, Published 4, November 1975, Crossland	15, 16
A	US, A, 3,242,038, Published 22, March 1966, Dallas et al.	15, 16
A	US, A, 3,239,478, Published 8, March 1966, Harlan, Jr.	15, 16
A	US, A, 4,096,203, Published 20, June 1978, St. Clair	15, 16
A	US, A, 3,935,338, Published 27, January 1976, Robertson et al.	15, 16
A	US, A, 3,828,367, Published 13, August 1974, Bourgeois	1-14
A	US, A, 3,600,250, Published 17, August 1971, Evans	1-14
A	US, A, 2,559,649, Published 10, July 1951, Little	11,12,13, 14
A	US, A, 3,976,530, Published 24, August 1976, Callan	13
A	US, A, 2,022,852, Published 3, December 1935, Galligan et al.	1
A	US, A, 3,932,328, Published 13, January 1976, Korpman	15-16

* Special categories of cited documents: ¹⁵

"A" document defining the general state of the art

"E" earlier document but published on or after the international filing date

"L" document cited for special reason other than those referred to in the other categories

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but on or after the priority date claimed

"T" later document published on or after the international filing date or priority date and not in conflict with the application, but cited to understand the principle or theory underlying the invention

"X" document of particular relevance

IV. CERTIFICATION

Date of the Actual Completion of the International Search ²	Date of Mailing of this International Search Report ²
January 2, 1980	15 JAN 1980

International Searching Authority ¹	Signature of Authorized Officer ¹⁰
ISA/US	Michael W. Ball

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